High-resolution electron time-of-flight apparatus for the soft x-ray region

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INTRODUCTION

The pulsed nature of synchrotron radiation (SR) emitted from electron or positron storage rings provided the basis for the development, about 25 years ago, of electron-TOF spectroscopy as an efficient, but relatively low resolution alternative to electrostatic or magnetostatic analysis. In the TOF technique, kinetic energies are determined by measuring flight times, typically up to several hundred nanoseconds, of electrons traveling a fixed distance between an interaction region and a detector. The method inherently relies on a coincidence between a timing pulse and an electron signal; background noise is suppressed and evenly distributed over the entire time spectrum, greatly simplifying data analysis. Another advantage of the TOF technique is that an entire electron spectrum can be collected simultaneously. In comparison, electrostatic analyzers, even with modern multichannel detection, collect only a portion of an electron spectrum at one time, which in some circumstances renders them susceptible to fluctuations in beam intensity and, in the gas phase, sample pressure. Simultaneous collection of an entire spectrum obviates these fluctuation effects and can increase the measurement efficiency of the electron-TOF method by a significant factor (up to 10^3 in the best of cases) relative to electrostatic analysis. A gas-phase time-of-flight (TOF) apparatus, capable of supporting as many as six electron-TOF analyzers viewing the same interaction region, has been developed to measure energy- and angle-resolved electrons with kinetic energies up to 5 keV. Each analyzer includes a newly designed lens system that can retard electrons to about 2% of their initial kinetic energy without significant loss of transmission; the analyzers can thus achieve a resolving power ($E/\Delta E$) greater than 10^4 over a wide kinetic-energy range. Such high resolving power is comparable to the photon energy resolution of state-of-the-art synchrotron–radiation beamlines in the soft x-ray range at the ALS, opening the TOF technique to numerous high-resolution applications. In addition, the angular placement of the analyzers, by design, permits detailed studies of nondipolar angular-distribution effects in gas-phase photoemission.

APPARATUS

A schematic of the experimental setup is shown in Fig. 1. An integral part of the apparatus is a differential-pumping section adapting the beamline pressure of less than 10^{-6} Pa to the chamber pressure during data collection of 4×10^{-3} Pa.

The vacuum chamber supporting the analyzers can be rotated about the x-ray beam (x axis) by ±90°, permitting collection of spectra at many different angles and thereby improving the precision and accuracy of angular-distribution measurements. An in-line rotational feedthrough decouples the chamber rotation from the differential-pumping section while the apparatus is under vacuum. Two gravitational sensors with an operating range of ±45.0° each

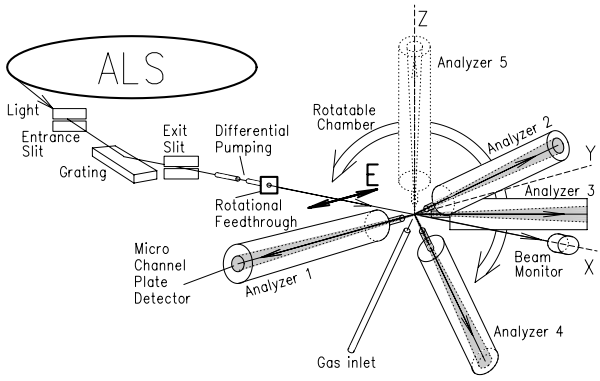

Figure 1. Experimental schematic of the electron time-of-flight system. Light from the ALS storage ring passes through beamline optics into a differential-pumping section. The chamber and analyzers can rotate around the photon beam for more accurate electron angular-distribution measurements.

determine the chamber rotation angle. The chamber is aligned with the x-ray beam along its rotational axis in order to avoid movement of the analyzers relative to the interaction region while rotating. A needle (30 mm long, 50 μm inner diameter, positioned less than 1 mm from the photon beam) directs an effusive jet of sample gas perpendicular to the x-ray beam. The interaction region viewed by the TOF analyzers runs about 3 mm along the x-ray beam and is defined by the beam size in the other dimensions. Sample gas enters the gas-inlet assembly through a manual leak valve. A 1000 *l* /s turbomolecular pump maintains the background pressure, with no sample gas flowing, of less than 10^{-5} Pa.

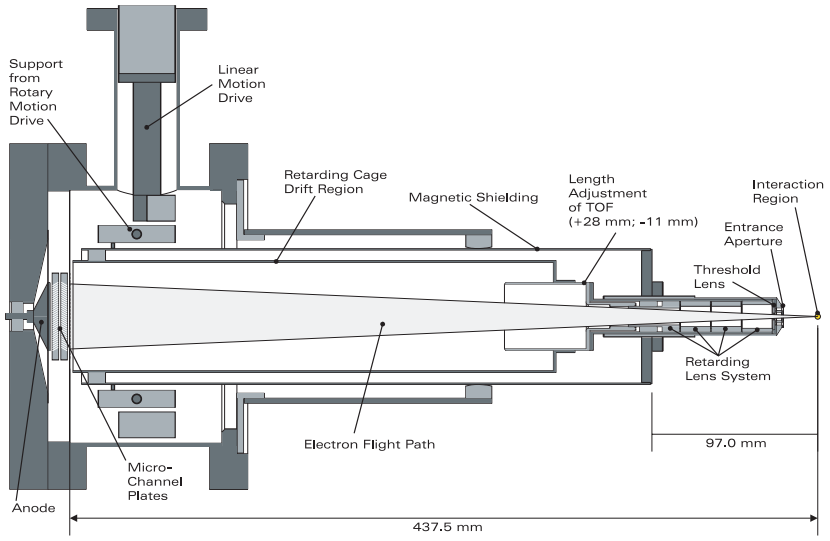

Figure 2. Cross section of an electron time-of-flight analyzer.

TOF ANALYZERS

Figure 2 shows a cross section of the electron-TOF analyzers. All analyzers are differentially pumped by a 50 or 80 *l* /s turbo pump to avoid pressure buildup near the MCPs, which have a recommended operating pressure of 10^{-4} Pa, significantly less than the chamber pressure during data collection. A straight electron flight path provides fundamental simplicity to the TOF technique, especially in analyzer design. The cylindrically symmetrical analyzers view the same interaction region with the 2 mm entrance apertures at a distance of about 20 mm. The apertures and needle are electrically grounded to maintain a field-free interaction region, an essential attribute for electron angular-distribution measurements. To optimize alignment to a common interaction region, the entrance aperture of each analyzer can be moved *in situ* ±3 mm along two axes perpendicular to the electron flight path, and the length of each analyzer can be adjusted *ex situ* +28 mm/-11 mm from its nominal length. A total flight path of 437.5 mm and minimum active areas on the MCPs of 41.9 mm diameter yields an angular acceptance of ±2.7°. This corresponds to a solid angle of 1/1800. Typical flight times are 738 and 73.8 ns for 1 and 100 eV electrons, respectively. With the time window of 328 ns between pulses in ALS two-bunch mode, a minimum kinetic energy of 5 eV is required for electrons to arrive at the detector before the next x-ray pulse reaches the interaction region. Electrons slower than 5 eV overlap with fast electrons created by the following bunch and complicate the spectra. If desired, the lens system can accelerate electrons to avoid these overlaps.

Figure 3 shows the front end of an analyzer, its lens system, and electron trajectories determined by ray tracing using SIMION software. In this simulation, the electrons begin in the interaction region with a kinetic energy of 505 eV, pass through the entrance aperture, then about 4.0 mm further pass through a second aperture with a diameter of 4.0 mm. The second aperture is usually at ground potential but can be biased with a negative 1–2 V to repel thermal electrons created in experiments with metal–vapor targets. Directly behind the second aperture is the first retardation element of the lens system at a potential of 80% (400 V) of the total retardation voltage of 500 V.

The next three elements have 91.5%, 97%, and 100% of the total voltage, respectively. The voltages are applied using a resistor cascade with the following approximate values: 15, 2.2, 1.0, and 0.56 MΩ, totaling 18.76 MΩ. After passing through the lens system, the electrons enter a field-free drift tube with a final kinetic energy of 5 eV. The drift tube extends to the MCP mounting. A coaxial magnetic-shield cylinder made of a Co–Netic AA alloy with a wall thickness of 0.76 mm surrounds the drift tube and reduces the magnetic field inside by a factor of up to 100, typically to about 4 mG. The shielding effect is less near the tube ends.

Figure 4 shows a neon spectrum taken at the 1*s*→3*p* resonance (867.1 eV) with flight time on the *x* axis and total counts on the *y* axis. The spectrum was collected for 200 s with a ring current of 20 mA. The entrance and exit slits were set at 50 μm, and the monochromator grating provided a bandpass of 650 meV, corresponding to a photon resolution of 1350. The analyzer was positioned parallel to the plane of the storage ring ($\theta = 0°$) and perpendicular to the photon beam ($\phi = 90°$). A retarding voltage of -750 V was applied to reduce the initial electron kinetic energies by 89%–97%. The fastest signal, at channel number 3930, is the prompt, created by fluorescent and scattered light, which indicates a timing resolution of 255 ps.

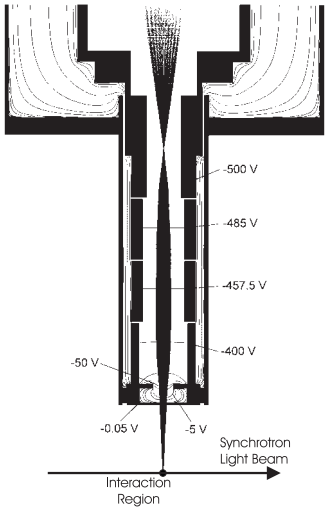

Figure 3. Cross section of the electron lens system right behind the entrance aperture of the analyzer. The trajectories of electrons with an initial kinetic energy of 505 eV passing through the retarding-lens system were calculated using SIMION software. An applied voltage of -500 V slows the electrons down to a final kinetic energy of 5 eV or about 1% of their initial kinetic energy.

APPLICATIONS

With this apparatus, angular distributions of valence photoelectrons showing effects due to higher-multipole interactions have been measured for the first time.[1,2] The experiments were performed on Ne 2*s* and 2*p* throughout the 250–1200 eV photon-energy range. Furthermore, it was possible to prove the breakdown of the independent-particle approximation throughout the same range.[3] Other measurements include the photon-energy dependence of ionization excitation in helium at medium energies (90–900 eV),[4] where we measured the photoionization-excitation-to-photoionization ratio for He^+ *nl* (*n*=2 – 6) and determined asymptotic high-energy ratios for He^+ *nl* (*n*=2 – 5). Current research projects include nondipole angular-distribution measurements in Xe and small molecules such as N_2 . We have observed a strong resonance-like increase in the value of the γ parameter about 60 eV above the N_2 1*s* ionization threshold, which causes an angular-distribution effect about 20 times more pronounced than predicted by theory for atomic nitrogen. Other projects include electron angular-distribution measurements of the Ar 2*s* satellite lines as well as the Ne valence lines in the Ne 1*s*→3*p* resonance region.

In conclusion, we have described the development of significantly improved electron-TOF analyzers for gas- phase samples and the design of an apparatus for angle-resolved-photoemission experiments to study dipole and nondipole angular-distribution effects. The new retarding-lens system increases the resolving power to up to 10^4 for high-kinetic-energy electrons. These improved electron-TOF analyzers are an alternative to electrostatic analyzers while maintaining the advantages of electron-TOF spectroscopy: constant background, large energy window, and independence of varying gas-target pressures and fluctuations in the beam intensity. A high-brilliance light source like the ALS with submillimeter beam sizes, combined

with this type of electron-TOF analyzer, provides state-of-the-art instrumentation for high-resolution angle-resolved photoelectron spectroscopy.

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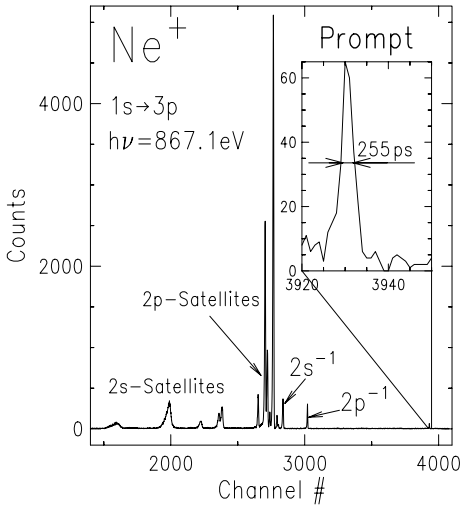

FIG. 4. Electron time-of-flight spectrum taken on the Ne 1*s*→3*p* resonance at 867.1 eV. The FWHM of the ‘‘prompt’’ (255 ps) determines the total timing resolution of the analyzer and electronics.

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